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Nano-anchor effect by anodic oxidation of aluminum sheets in joining by electrodeposition

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Abstract

Two anodized and stacked aluminum sheets were adhered by electrodeposition of copper. The anodizing conditions were varied and the adhesion strength (interface shear strength) was investigated by lap shear tests. The interfacial shear strength depended on the conditions of the anodic oxidation and reached >60 MPa when the aluminum sheets was anodized in H_3PO_4 before electrodeposition of copper. An anodic aluminum oxide film was formed by anodic oxidation (anodization) of the aluminum sheets and had porous structure with straight nanopores perpendicular to the surface. Hence, anodized Al sheets were strongly adhered by Cu electrodeposition because of nano-anchor or interlocking effect between the porous structure and electrodeposited copper. The proposed joining technique through electrodeposition may be useful because it can provide electrically conductive joints at room temperature and ambient pressure.

Keywords: Aluminum sheets; Joining; Electrodeposition; Interfacial shear strength

1. Introduction

Joining techniques of metallic dissimilar sheets to form layers or laminates, such as rolling followed by annealing [1], friction stir welding [2], laser welding [3], ultrasonic spot welding [4] and explosive welding [5] have been

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developed so far. In these processes, a thermal or diffusion process is required to bond the layers because bonding at room temperature is suppressed by rigid oxide films on the surface of metallic materials. However, thermal or diffusion processes inevitably form chemical compounds at the interfaces between the dissimilar layers, which leads to premature fracturing or a reduced ductility [6]. It is desirable to develop a new manufacturing process for metallic layers without using a thermal process.

There are three mechanisms for the adhesion of metallic sheets: physical (diffusional) bonding, chemical bonding, and mechanical interlocking (anchor effect). Mechanical interlocking is an alternative for strongly adhering metallic sheets without thermal processes.

An anodic aluminum oxide film with nanoporous structure is formed by the electrochemical oxidation (or anodization) of aluminum. After the study by Masuda et al. [7] which showed highly ordered nanohole arrays in anodic aluminum oxide and their application, many studies have been performed on the ordered nanoporous anodic aluminum oxide films [8, 9]. Recently, it has been found that an electrodeposited copper film adheres strongly to anodized aluminum [10, 11]. The enhanced adhesion is due to the interlocking by the penetration of the electrodeposited copper into the nanopores of anodic aluminum oxide films. Thus, anodized aluminum sheets can be adhered strongly by copper electrodeposition because of the nano-anchor effect (Fig. 1(a)).

We have reported the results of the preliminary experiments where two anodized aluminum sheets were adhered by electrodeposition of copper (Fig. 1(b)) with interfacial shear strength of >60 MPa [12]. In the present paper, the conditions of anodic oxidation of aluminum sheets were varied and adhesion strength (interface shear strength) was investigated by lap shear tests.

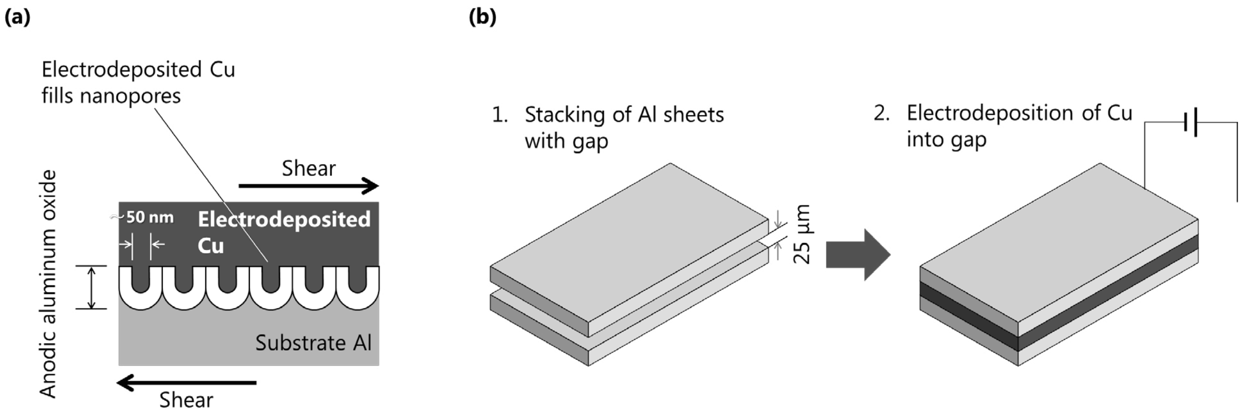

Fig. 1. Schematic illustrations for (a) nano-anchor effect by nanoporous structure in anodic aluminum oxide and (b) joining of aluminum sheets by electrodeposition of copper.

2. Experimental procedures

Rectangular sheets made of pure aluminum were prepared by cutting purchased aluminum sheets (Nilaco, Japan). The dimension of the sheets was 30 mm long, 4 mm wide and 1.5 mm thick. One surface (30 mm × 4 mm) of the aluminum sheet was machined to obtain a pentagonal cross section with an edge thickness of 1.4 mm [12]. Some of the aluminum sheets were then anodized under conditions where the electrolyte was 0.3 M H_3PO_4 or 0.3 M H_2CrO_4. The anodizing voltage was 30 V and the anodizing conduction time was 20–60 min. A SUS316 sheet (Nilaco, Japan) was used as a cathode in the anodization.

Followed by the anodization, electrodeposition of copper between the two stacked aluminum sheets was performed. In other words, the gap between the two aluminum sheets was filled by copper electrodeposition. The conditions for copper electrodeposition were typical as summarized in Table 1. Two anodized sheets were stacked with a gap of 25 μm between them. The area for copper electrodeposition was 20 mm × 4 mm. The aluminum sheets, consisting of two anodized aluminum sheets, was set as a cathode and a copper sheet was set as an anode in a sulfate bath. Direct current was used for the electrodeposition for simplicity.

Table 1. Conditions for electrodeposition of copper between two stacked aluminum sheets.

Bath component	$CuSO_4 \cdot 5H_2O$ 220 g/L H_2SO_4 60 g/L NaCl 0.1 g/L Polyethyleneglycol 0.5 g/L
Bath volume	250 mL
Current density	1.88 A/dm^2
Conduction time	24 hours
Temperature	298 K
Distance between anode and cathode	50 mm

Lap shear tests were conducted on the aluminum sheets joined by the electrodeposition of copper at room temperature to investigate the interface shear strength according to modified JIS K 6850 [13]. An overlap length was 20 mm and a sample width was 4 mm.

The microstructures of the samples (including cross sections) were observed with a field emission scanning electron microscope (FE-SEM, SU-6600, Hitachi High-Technologies Corporation, Japan) equipped with an energy-dispersive X-ray spectrometer (EDXS, XFlash 5010, Bruker AXS, Germany).

3. Results and discussion

3.1. Electrodeposition between stacked aluminum sheets

Copper was preliminarily electrodeposited between the stacked aluminium sheets without grinding to form pentagonal cross sections. The two sheets were joined at the edges; however, as schematically shown in Fig. 2(a), the cross-sectional observation revealed that the large cavity was inevitably formed, probably due to the insufficient supply of copper ion from bulk electrodeposition bath. Therefore we adopted the pentagonal cross sections of the stacking of the sheets so that the copper ion was successively supplied during the electrodeposition. As a result, most of the gap was filled and the robust joint without fracturing during handling was obtained. Homogeneous electrodeposition in a gap, actually, will be one of the subjects to be solved for practical application.

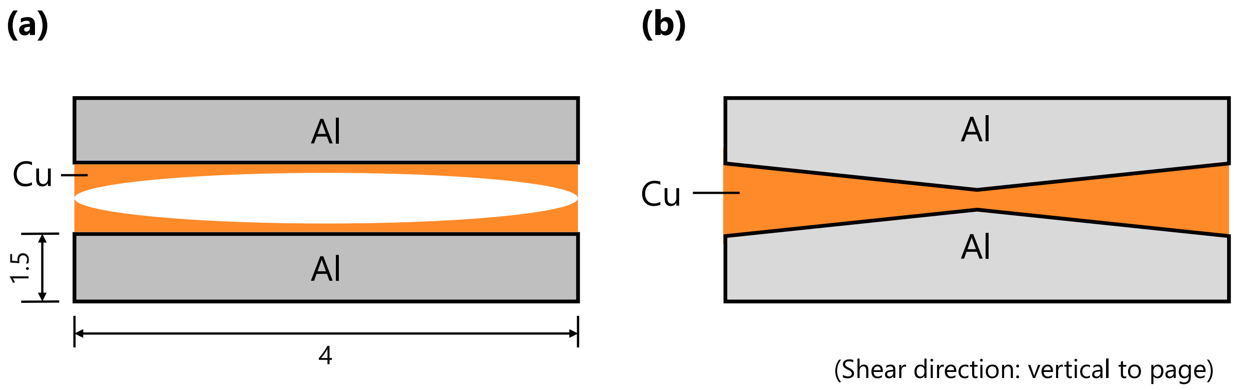

Fig. 2. Schematic illustrations of macroscopic features of (a) rectangular and (b) pentagonal cross sections of two aluminum sheets joined by electrodeposition of copper.

3.2. Anodized aluminum oxides

The surface microstructures of aluminum anodized in 0.3 M H_3PO_4 were observed with an SEM and are shown in Fig. 3. The SEM observations of the anodized aluminum sheets often suffered from halation when a high acceleration voltage (20 kV) of electron beam was applied; therefore these images were observed under a low acceleration voltage of 2.0 kV. The decrease in the electrical conductivity, which is a main reason for the halation, is likely due to the formation of lowly-conductive anodic aluminum oxide on the surface of aluminum. Fine pores was uniformly formed in aluminum oxide anodized in H_3PO_4. The pore diameter and porosity was approximately 50 nm and 33%, respectively, regardless of the anodization time adopted here. Although the cross sections of the as-synthesized anodic aluminum oxide were not observed, it is expected that the pores are almost vertical to the surface and that the average depth of the pores increases with the anodization time.

SEM images of surface microstructures of aluminum anodized in 0.3 M H_2CrO_4 are shown in Fig. 4. Nanopores were successfully formed in the anodic aluminum oxide film anodized in 0.3 M H_2CrO_4; however, the approximate pore diameter and porosity were 20 nm and 10%, respectively, which are smaller than those in anodic aluminum oxide formed in 0.3 M H_3PO_4. These observations indicate that the pore formation in anodic aluminum oxide was more facilitated by anodizing in H_3PO_4 than by anodizing in H_2CrO_4.

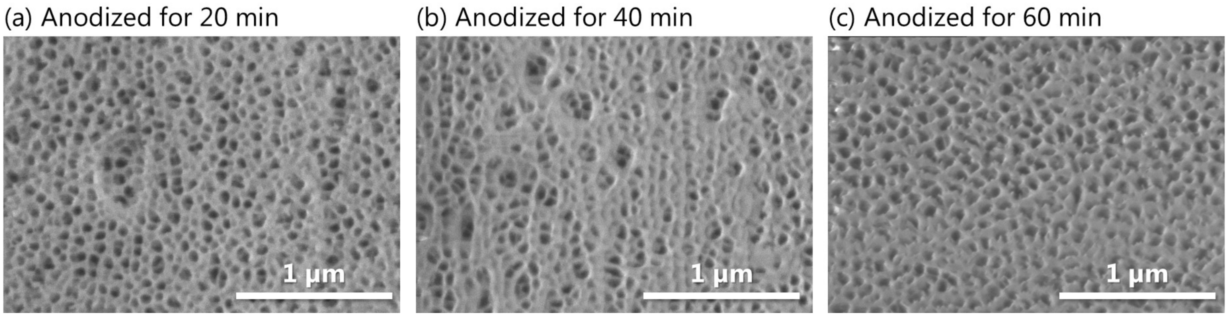

Fig. 3. Scanning electron microscopic images of surfaces of aluminum sheets anodized in 0.3 M H_3PO_4 for (a) 20, (b) 40 and (c) 60 min.

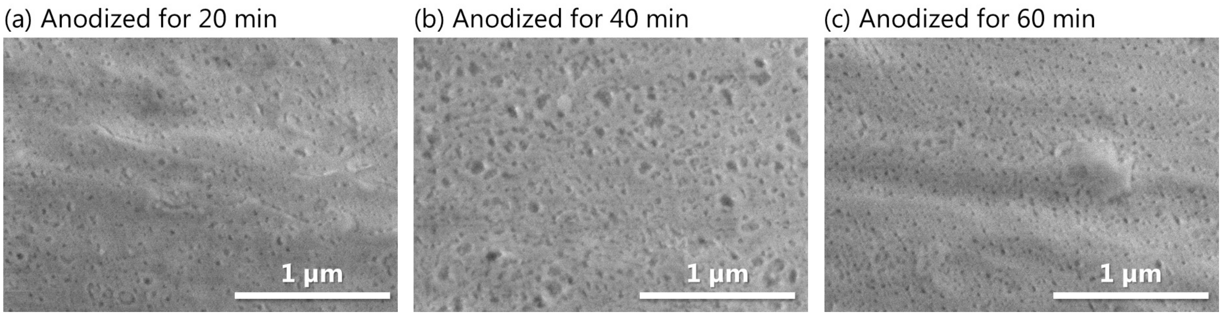

Fig. 4. Scanning electron microscopic images of surfaces of aluminum sheets anodized in 0.3 M H_2CrO_4 for (a) 20, (b) 40 and (c) 60 min.

3.3. Lap shear tests

The results for lap shear tests are summarized in Fig. 5(a). Without anodic oxidation, the tensile shear strength of the interface between stacked aluminum sheets was 1.5 MPa. However, the anodized aluminum sheets were bonded with improved tensile shear strength above 4 MPa, except for the aluminum sheets treated by 20-min anodization in H_2CrO_4. Most of the samples fractured at the interface between aluminum and copper during the lap tests; however, the samples treated by 40-min and 60-min anodization in H_3PO_4 fractured at the void in the electrodeposited copper, which inevitably forms even the pentagonal cross sections (Fig. 2) are adopted. These two cases showed tensile shear strength higher than 7 MPa; thus, the aluminum/copper interface in these samples are significantly stronger than the other samples.

To clarify the shear strength of the aluminum/copper interface, additional lap shear tests where only bonding area of aluminum/copper was reduced to 1.5 mm × 1.2 mm were carried out on the bonded aluminum sheets anodized for

40-min and 60-min in H_3PO_4. In this configuration with the reduced interfacial area, the void formation in the electrodeposited copper was limited, and as a result, fractures resulted at the aluminum/copper interface and the shear strengths of the aluminum/copper interface were above 60 MPa (Fig. 5b). Much higher interfacial shear strength than the aluminum/copper interface without anodization was attained by the 40-min and 60-min anodization of aluminum sheets in H_3PO_4.

These lap shear tests, although the adopted bonding area was not united, suggest that anodization of aluminum sheets significantly strengthens the shear strength of the aluminum/copper interface. The nanoporous feature of anodic aluminum oxide films seems closely related to the strengthening of the interface.

Cross-sectional SEM images of the aluminum/copper interfaces are shown in Figs. 6 and 7. Copper was electrodeposited in the nanopores, forming copper nanorods array in anodic aluminum oxide. The filling of nanopores by electrodeposited copper is likely to cause nano-anchor effect and to be a main source of the improved interfacial shear strength. Anodization in H_2CrO_4 resulted in the nanorods array with smaller diameter and density than those in the anodic aluminum oxide processed in H_3PO_4, suggesting that larger pore density (porosity) and larger pore size is required for better nano-anchor effect.

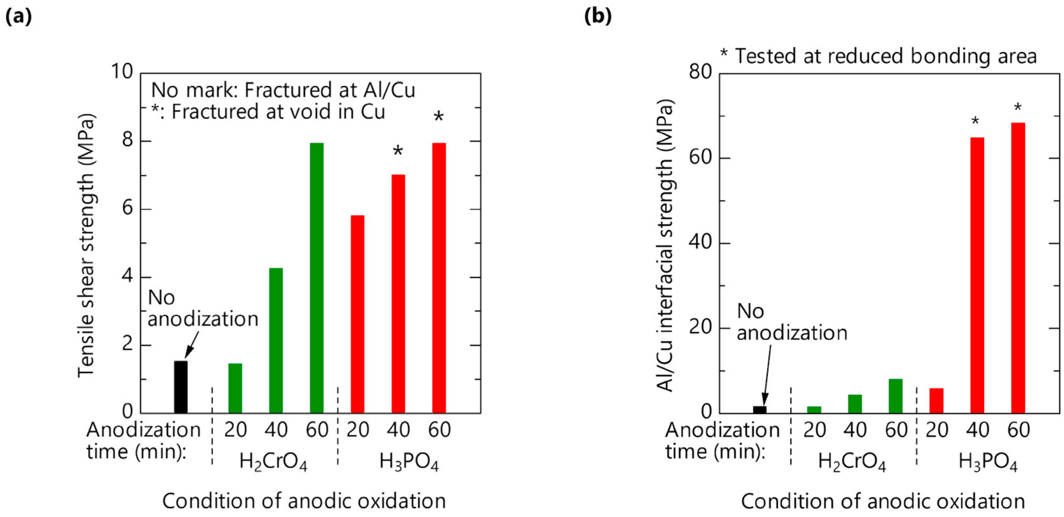

Fig. 5. Results of lap shear tests of aluminum joined by electrodeposition. Graph (a) shows results of tests under constant bonding area, while (b) shows results of tests under different bonding area to elucidate Al/Cu interfacial shear strength.

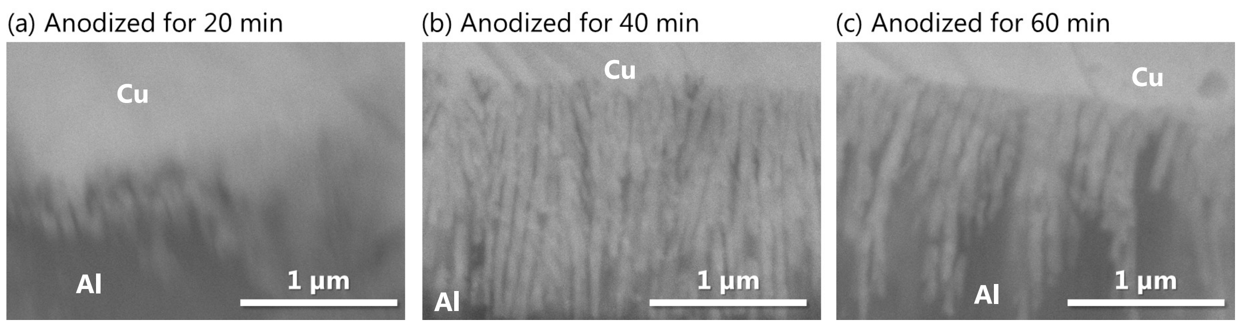

Fig. 6. Cross sectional scanning electron microscopic images of anodized aluminum sheets/electrodeposited copper. Anodization was conducted in H_3PO_4.

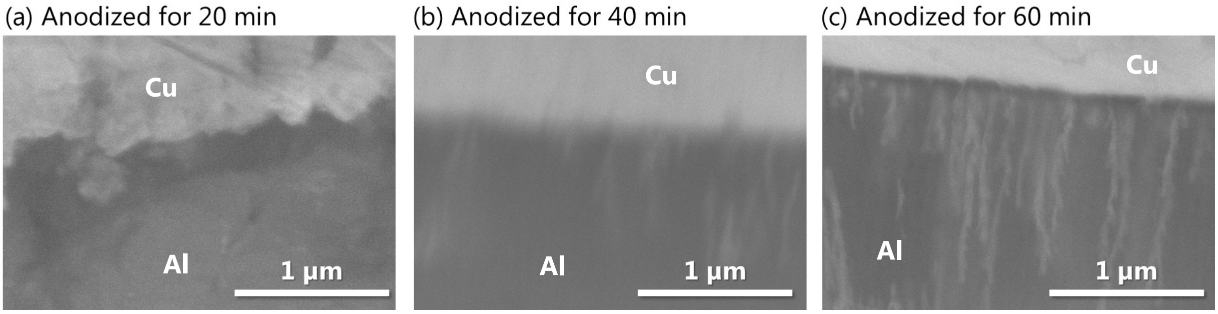

Fig. 7. Cross sectional scanning electron microscopic images of anodized aluminum sheets/electrodeposited copper. Anodization was conducted in H_2CrO_4.

Acknowledgements

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